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THE PRESENCE OF ^{146}Sm IN THE EARLY SOLAR SYSTEM AND IMPLICATIONS FOR ITS NUCLEOSYNTHESIS

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ABSTRACT

The presence of the *p*-process nucleus ^{146}Sm (mean life, $\bar{\tau} = 149 \times 10^6$ yr) in the early solar system and its *in situ* α-decay into ^{142}Nd is demonstrated by the correlation of ^{142}Nd/^{144}Nd with ^{144}Sm/^{144}Nd in two meteorites which have a large range in ^{144}Sm/^{144}Nd in their constituent mineral phases. Clear excesses of ^{142}Nd/^{144}Nd, relative to the solar system value, are present in high Sm/Nd phases and a clear deficit of ^{142}Nd/^{144}Nd is observed in one sample with low Sm/Nd. The inferred abundance of ^{146}Sm/^{144}Sm is 0.008 at the time of the last equilibration of each meteorite at 4.47 AE ago, which yields ^{146}Sm/^{144}Sm $\sim$ 0.015 at the time of formation of the solar system, 4.56 AE ago. These results confirm the presence of ^{146}Sm and provide a well-defined initial abundance for ^{146}Sm. The abundance of ^{146}Sm is compatible with the *p*-process production rate estimates but not with the production rate for ^{146}Sm based on a photodisintegration model for the production of *p*-process nuclides.

Subject headings: abundances — meteors and meteorites — nucleosynthesis

I. INTRODUCTION

Proton-rich isotopes are dominantly produced by the *p*- or γ-processes (Burbidge *et al.* 1957, hereafter B^{2}FH; Cameron 1957; Audouze and Truran 1975; Woosley and Howard 1978). Isotopes ascribed to this process are less abundant (typically by a factor of 10–1000) than those produced by neutron-capture processes. The measured solar system abundances of the proton-rich isotopes (cf. compilations by Cameron 1982 and Anders and Ebihara 1982) have been modeled by nucleosynthetic calculations (B^{2}FH; Audouze and Truran 1975; Woosley and Howard 1978). Some *p*-process radioactive nuclei, which are now extinct, have mean lives long enough to have been possibly present in the early solar system (^{92}Nb, $\bar{\tau} = 53 \times 10^6$ yr; ^{146}Sm, $\bar{\tau} = 149 \times 10^6$ yr). Since the mean life of ^{146}Sm is of the same order as the time interval of formation of the planets in our solar system, it would be reasonable to expect its presence in solar system planetary bodies. This would complement the presence of shorter lived nuclides in the early solar system produced by other processes (e.g., ^{26}Al, ^{53}Mn, ^{107}Pd; cf. review by Wasserburg 1985) and would possibly have direct chronological implications for a specific nucleosynthetic process (Audouze and Schramm 1972). Demonstration of the existence of ^{146}Sm and determination of its abundance depends on the observation of variations in the abundances of the ^{142}Nd daughter. Because another pair of isotopes of these two elements (parent ^{147}Sm, $\bar{\tau} = 153 \times 10^9$ yr, daughter ^{143}Nd) has been used as an absolute chronometer, it may be possible to combine the long-lived chronometer (^{147}Sm-^{143}Nd) and the potential short-lived one (^{146}Sm-^{142}Nd).

The main issues to be considered are whether or not (a) ^{146}Sm was present in the early solar system and can be used as a chronometer and (*b*) the current models of nucleosynthetic processes and calculated production rates are consistent with the abundance of ^{146}Sm inferred from experiments. As both ^{146}Sm and ^{144}Sm are assumed to be generated by the *p*- or γ-process, it is customary to relate the inferred abundance of ^{146}Sm to that of the stable ^{144}Sm. In previous experiments, claims have been made for the presence of ^{146}Sm with an inferred abundance of ^{146}Sm/^{144}Sm at 4.56 AE ranging from 0.005 (Lugmair and Marti 1977; Lugmair *et al.* 1983) to 0.013 (Jacobsen and Wasserburg 1984). We demonstrate that for two meteorites, Ibitira and Morristown, chosen to contain minerals with large Sm/Nd chemical fractionation, a correlation can be found between Sm/Nd and excesses and deficits in ^{142}Nd/^{144}Nd.

Ibitira is the only known unbrecciated basaltic eucrite. The presence of vesicles supports an igneous origin (Wilkening and Anders 1975; Steele and Smith 1976), although the texture indicates some recrystallization (Steele and Smith 1976). Its mineralogy is close to that of a basalt: 60% pyroxene, 30% plagioclase, and 0.1% phosphate, plus other minor phases with almost no Sm or Nd. This meteorite has been studied for U-Th-Pb (Chen and Wasserburg 1985) and contains fission Xe from ^{244}Pu and a ^{129}Xe excess, possibly from ^{129}I (Wasserburg *et al.* 1977). The second meteorite is the class 3A mesosiderite Morristown (Floran 1978). It has a brecciated texture with a matrix composed of intimate intergrowths of silicate (50%) and FeNi metal (50%). Clasts (cm to mm in size) are embedded in the matrix, half of them composed mainly of silicates and the others mainly of FeNi metal. The nonmetallic fraction is comprised of poikilitic orthopyroxene (80%), plagioclase (14%), and phosphate ($\sim$3%). The formation of mesosiderites is not well understood, but requires the segregation of metal-rich zones during the differentiation of small planets. It is also possible that an impact produced these brecciated meteorites.

For both samples, it was possible to analyze three phases which contain the rare earth elements: plagioclase, phosphate, and pyroxene. The existence of three independent phases with a large variation in Sm/Nd is critical in determining whether the correlation of the ^{142}Nd/^{144}Nd with Sm/Nd represents an isochron. The chemical fractionation factors, $f_{\mathrm{Sm/Nd}}$, relative to the mean Sm/Nd for chondrites (see Table 1), vary from -0.22 to 0.49 for Ibitira, and from -0.64 to 0.81 for Morristown. These large ranges of $f_{\mathrm{Sm/Nd}}$ permit an accurate estimate of the existence of ^{146}Sm in the early solar system.

II. RESULTS

Mass spectrometric data for Nd were acquired in three ways and the results were shown to be consistent.

1. Measurement of the full sequence of seven Nd isotopes: variations in Nd isotope abundances are calculated as $\epsilon_i = [({}^{i}\mathrm{Nd}/{}^{144}\mathrm{Nd})/({}^{i}\mathrm{Nd}/{}^{144}\mathrm{Nd})_{\mathrm{CHUR}} - 1] \times 10^4$, using the ^{142}Nd/^{146}Nd ratio for the normalization of mass-dependent isotope fractionation. This corresponds to our standard convention. However, because ^{142}Nd shows anomalies, this normalization results in apparent shifts for the ϵ-values of the remaining isotopes. Since the isotope fractionation is small, these apparent shifts in ϵ-values are a linear function of the mass difference. We calculated a regression line through the apparent shifts: ϵ_{144}, ϵ_{145}, ϵ_{146}, ϵ_{148}, and ϵ_{150}, for unspiked samples. The deviations of ϵ_{142} (excess or deficit) from this regression line yielded the values of ϵ_{142} in Table 1.

2. With the same sequence of seven isotope measurements, we corrected the mass-dependent fractionation using ^{144}Nd/^{146}Nd for normalization. The results are fully consistent with the first calculation, and only results from (1) are listed in Table 1.

3. In order to improve the ^{142}Nd measurement, we also measured cycles of only three isotopes (^{142}Nd, ^{144}Nd, and ^{146}Nd) and corrected the mass-dependent fractionation, using the ratio ^{144}Nd/^{146}Nd.

TABLE 1

ANALYTICAL RESULTS

Sample[a]	^{144}Nd (nmoles g^{-1})	^{144}Sm (pmoles g^{-1})	$f^{\mathrm{Sm/Nd}}_{\mathrm{CHUR}}$	ϵ(142)
		Ibitira		
WR (172)	9.0311±16	359.0±5	−0.0143	0.18±0.41
				0.22±0.30
PX + PH (60)	6.6137±17	310.1±3	0.1626	0.20±0.50
				0.77±0.58
	...	...	...	0.42±0.36
PX (64)	3.8699±6	233.0±6	0.4928	2.27±0.36
	...	...	...	1.89±0.26
				2.09±0.24[b]
PL + PH (16)	12.880±2	424.5±1.3	−0.1828	−0.09±0.26
PL (19)	3.1597±5	99.7±1	−0.2175	0.02±0.34
	...	...	...	−0.07±0.47[b]
PH	0.015226±3[c]	0.5054±10[c]	−0.1770	−0.50±0.62
		Morristown		
PH	0.20823±4[c]	11.46±7[c]	0.3651	1.63±0.52
				1.04±0.31
				1.72±0.28
				1.75±0.28[b]
WR				
residue (156)	0.068268±15	3.724±7	0.3524	1.71±0.59
PL (191)	0.091585±9	1.323±2	−0.6419	−1.15±0.35
PX (492)	0.011095±5	0.809±5	0.8086	2.04±1.26

NOTE.—Each entry represents separate individual runs. Uncertainties (2 σ) correspond to last significant figures. $f^{\mathrm{Sm/Nd}}_{\mathrm{CHUR}}$: chemical fractionation factor relative to the Sm/Nd in a CHondritic Unfractionated Reservoir (CHUR, DePaolo and Wasserburg 1976; Jacobsen and Wasserburg 1980). $f^{\mathrm{Sm/Nd}}_{\mathrm{CHUR}} = ({}^{144}\mathrm{Sm}/{}^{144}\mathrm{Nd})/0.04033 - 1$. Results of ^{147}Sm-^{143}Nd for Ibitira: $T = 4.46 \pm 0.03$ AE; $({}^{143}\mathrm{Nd}/{}^{144}\mathrm{Nd})_I = 0.506090 \pm 38$; $\epsilon^I_{143} = 1.55 \pm 0.76$ ϵu; for Morristown: $T = 4.47 \pm 0.02$ AE; $({}^{143}\mathrm{Nd}/{}^{144}\mathrm{Nd})_I = 0.506104 \pm 33$; $\epsilon^I_{143} = 2.09 \pm 0.65$ ϵu.

[a] WR = whole rock; PX = pyroxene; PH = phosphate; PL = plagioclase. Numbers in parentheses are sample weights (mg).

[b] Isotopic ratios obtained with cycles of three isotopes (^{142}Nd, ^{144}Nd, ^{146}Nd) corrected for mass-dependent fractionation with ^{146}Nd/^{144}Nd = 0.724134.

[c] Nanomoles of ^{144}Nd and picomoles of ^{144}Sm in the leaches, assumed to be controlled by the phosphate for the rare earths.

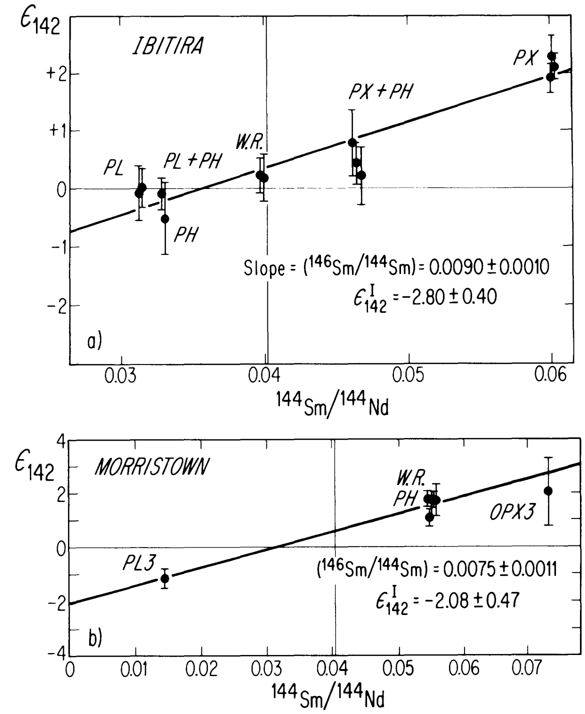

FIG. 1.—^{146}Sm-^{142}Nd evolution diagrams for the Ibitira eucrite (*top*) and the Morristown mesosiderite (*bottom*). The relatively old age and the large range in Sm/Nd allowed the preservation of deficits and excesses in ^{142}Nd for phases with low and high Sm/Nd, respectively, relative to the chondritic (CHUR) reservoir. The present-day composition of CHUR is provided by the intersection of $\epsilon_{142} = 0$ and ^{144}Sm/^{144}Nd = 0.0403 as shown.

Well-defined absolute ages were obtained from ^{147}Sm-^{143}Nd internal isochrons. These ages are 4.46 ± 0.03 AE for Ibitira and 4.47 ± 0.02 AE for Morristown (Table 1). The ages are old enough to permit the identification of the decay products of ^{146}Sm if it were present. All uncertainties in this *Letter* are at the 95% confidence level. The initial ^{143}Nd/^{144}Nd (ϵ^I_{143} at 4.46 and 4.47 AE) isotopic compositions indicate that these meteorites were formed from precursor material which had been fractionated relative to bulk solar system values.

Ibitira.—A specimen of 170 mg was crushed and dissolved for whole rock (WR) analysis. Another chunk of rock was crushed for mineral separates. A first magnetic separation at coarse grain size (~150 μm) gave a pyroxene-rich (PX + PH) and a plagioclase-rich (PL + PH) fraction. To remove the phosphate (PH) inclusions and obtain pure mineral phases (PX, PL), the samples were crushed to finer size (< 40 μm), and a more careful mechanical separation (magnetic + density) was carried out. The two best separates (PX and PL) were dissolved without any leaching. The phosphate analysis (PH) was performed on the leach (with weak nitric acid) of a phosphate-rich fraction containing plagioclase. For the three phases analyzed, there is full agreement between the modal mineral proportions and the Nd and Sm mass balance between these minerals. The measurements of ϵ_{142} show a correlation with the ^{144}Sm/^{144}Nd ratio (Fig. 1 [*top*]). This correlation is interpreted as an isochron due to the *in situ* decay of ^{146}Sm.

From this diagram, we conclude that ϵ_{142} is correlated with ^{144}Sm/^{144}Nd and calculate ^{146}Sm/^{144}Sm = ^{142}Nd*/^{144}Sm = 0.0090 ± 0.0010 (* = excess) at the time of closure of the Sm-Nd system in Ibitira, which is presumed to be the time of crystallization. The regression line passes close to the present-day point for CHUR (^{144}Sm/^{144}Nd = 0.0403; ϵ_{142} = 0). However, the small positive ϵ_{142} shift is consistent with the ϵ^I_{143} of Ibitira, which indicates fractionated Sm/Nd for the parent reservoir.

Morristown.—A chunk of a silicate-rich portion was crushed for mineral separates. The metal portions were removed during crushing. The whole rock fine powder (<40 μm) was first leached with weak nitric acid (the leach is dominated by the phosphate, fraction PH, Table 1), and then dissolved (WR residue, Table 1). The leach and residue show the same Sm/Nd and Nd isotopic composition. The phosphate and the WR residue show a clear excess of ^{142}Nd. As expected, the vast majority of Nd and Sm (~98%) is resident in the phosphate. As the amount of Nd was too small (<1 ng) in the orthopyroxene and plagioclase separates from this chunk to get a precise analysis for ϵ_{142}, we crushed a larger piece (~33 g), and mechanically separated orthopyroxene and plagioclase fractions. Both fractions were gently leached with dilute warm aqua regia to remove traces of phosphate. The plagioclase, with an extremely low Sm/Nd, shows a clear deficit in ^{142}Nd, while the pyroxene, with a large positive f value shows a large excess of ^{142}Nd (Fig. 1 [*bottom*]). The results from these four Morristown samples (WR residue, PH, PL, and PX) with very different Sm/Nd exhibit a well-defined correlation of Sm/Nd with both excesses and deficits in ^{142}Nd. The slope of this correlation implies ^{146}Sm/^{144}Sm = 0.0075 ± 0.0011 at the time of crystallization. This correlation line shows a small, resolvable shift from the present-day CHUR indicating a source with fractionated Sm/Nd. This fractionation, which yields a positive ϵ^I_{142} is consistent with the fractionation of Sm/Nd in the parent material in which this meteorite formed, as implied by the initial ^{143}Nd/^{144}Nd (see note to Table 1).

III. DISCUSSION

The data present well-defined effects for ^{142}Nd, both excesses and deficits, which are correlated with ^{144}Sm/^{144}Nd. These data demonstrate the earlier presence of the extinct ^{146}Sm. We interpret the excesses and deficits of ^{142}Nd as reflecting the formation of crystals at the time when sufficient ^{146}Sm was still present. From the slope of the correlation lines, we calculate ^{146}Sm/^{144}Sm at the time of crystallization of 0.0090 ± 10 and 0.0075 ± 11 for Ibitira and Morristown, respectively. The ages at which these abundances were present are determined by the decay of ^{147}Sm and are 4.46 ± 0.03 AE and 4.47 ± 0.02 AE, respectively (Prinzhofer, Papanastassiou, and Wasserburg 1989). The two meteorites have identical ^{147}Sm-^{143}Nd ages and identical initial ^{146}Sm/^{144}Sm abundances. These data imply a uniform ^{146}Sm/^{144}Sm in diverse objects and suggest the possibility that ^{146}Sm-^{142}Nd may be useful as a chronometer which can encompass the time of planetary formation. If we consider T = 4.56 AE as the time of formation of the solar system, we calculate the initial ^{146}Sm/^{144}Sm = 0.017 ± 0.002 and 0.014 ± 0.002 from Ibitira and Morristown, respectively. We conclude from our data that there is clear evidence for the presence of ^{146}Sm within the solar system, with ^{146}Sm/^{144}Sm ~ 0.015 at 4.56 AE. This implies that about 5×10^{-4} of the ^{142}Nd found in CHUR was also produced by the α-decay of ^{146}Sm in the solar system.

Early data on ^{142}Nd by Notsu *et al.* (1973) from the Juvinas achondrite claiming large ^{142}Nd excesses were shown by Lugmair, Scheinin, and Marti (1975) to be incorrect. Within 2 σ errors, the latter workers deduced a ^{146}Sm/^{144}Sm ratio for Juvinas at the time of crystallization of 0.0054 ± 0.0072, indistinguishable from zero. The poor resolution of this value comes from the small range of Sm/Nd fractionation found in this meteorite. Careful work on the achondrites Angra dos Reis (ADOR; Lugmair and Marti 1977), Moama, and Angra dos Reis (Jacobsen and Wasserburg 1984) provided evidence of extinct ^{146}Sm. A discrepancy exists for the ^{146}Sm/^{144}Sm ratio for ADOR, dated at 4.564 ± 0.037 AE (Jacobsen and Wasserburg 1984). The reported ratios are 0.0047 ± 0.0023 by Lugmair and Marti (1977) and 0.0118 ± 0.0032 by Jacobsen and Wasserburg (1984). The reported ratio for Moama is 0.0041 ± 0.0013, which, considering the younger age for this meteorite, gives 0.008 ± 0.003 at 4.56 AE. Lugmair *et al.* (1983) also found very large excesses of ^{142}Nd and ^{143}Nd in Allende acid-resistant residues. These anomalies are not correlated with Sm/Nd, and were interpreted as due to implantation into pre-existing thin coatings of the silicate grains, due to recoil from decay of the parent nuclei. They calculated a ^{146}Sm/^{144}Sm = 0.0045 ± 0.0005 from the ^{142}Nd*/^{143}Nd* ratio, after having demonstrated normal isotopic composition for Sm and chondritic Sm/Nd ratio for the sample. No absolute time from ^{147}Sm-^{143}Nd can be assigned to this estimate of ^{146}Sm/^{144}Sm.

Comparing these previous estimates to the present data, we obtain the same ^{146}Sm/^{144}Sm ratio at 4.56 AE as Jacobsen and Wasserburg (1984) obtained for ADOR. This ratio is about twice as large as that calculated by Lugmair and Marti (1977) for ADOR, and by Jacobsen and Wasserburg (1984) for Moama. To check this discrepancy, we measured a new pyroxene separate of ADOR. The new data lie exactly on the two isochrons ^{147}Sm-^{143}Nd and ^{146}Sm-^{142}Nd defined in Figures 5 and 6 of Jacobsen and Wasserburg (1984). The pyroxene, with a Nd isotopic composition of CHUR, does not show any anomaly in ^{142}Nd. We believe that this agreement establishes the compatibility of our data with those of Jacobsen and Wasserburg (1984) for ADOR. The discrepancy with the other results may reflect heterogeneity of ^{146}Sm/^{144}Sm in the solar nebula, or more plausibly, as seems to be the case of ADOR, experimental difficulties for some of the data.

The relation between ^{146}Sm/^{144}Sm in the solar system at 4.56 AE and the production rate of ^{146}Sm relative to ^{144}Sm, $P_{146/144}$, depends on the nucleosynthetic process and on the time dependence of element production in the galaxy. Following the formalism of Schramm and Wasserburg (1970), we obtain

$$(^{146}\mathrm{Sm}/^{144}\mathrm{Sm})_{\odot} = P_{146/144}\left[\frac{1-X}{\lambda_{146}T^*} + X\right]e^{-\lambda_{146}\Delta}\,, \quad (1)$$

where X is the fraction of ^{146}Sm introduced in the system in a single late spike; λ_{146}, 6.73×10^{-9} yr^{-1}; $T^* = T\langle p\rangle/p(T)$ with T = total duration; $\langle p\rangle$ = mean production rate; $p(T)$ = last production rate; Δ, the time delay between the last nucleosynthetic "p" process contribution to the solar system and condensation in the solar system.

We will consider the two extreme cases: (*a*) all the ^{146}Sm and ^{144}Sm are introduced as a late spike (X = 1), and (*b*) there is no late spike (X = 0), but a continuous nucleosynthesis. In both cases, we will assume that formation of solid bodies in the solar system begins immediately after the last nucleosynthetic event ($\Delta = 0$).

In the first case, assuming that nucleosynthesis occurred at 4.56 AE ago, we obtain, at that time: $(^{146}\mathrm{Sm}/^{144}\mathrm{Sm})_\odot = P_{146/144}$, which implies $P_{146/144} \approx 0.015$ from our results. In the second case, at 4.56 AE, equation (1) yields $(^{146}\mathrm{Sm}/^{144}\mathrm{Sm})_\odot = P_{146/144}(\lambda T^*)^{-1}$. T^* can be estimated as ~ 7 AE for a uniform nucleosynthesis model, or as ~ 1 AE for an increasing production rate toward the time of isolation of our solar system. For these cases, $P_{146/144}$ would fall between 0.7 and 0.1.

Different calculations have been proposed by previous authors to estimate the production ratio $P_{146/144}$ using elemental and isotopic systematics from cosmic abundances. Audouze and Schramm (1972) estimated $P_{146/144}$ using a logarithmic interpolation of the *p*-process isotope abundances in the range $A = 149$–156. Their estimate of the ^{146}Sm production rate is $0.35 < P_{146/144} < 0.6$. Again using this approach, from a comparison with the production rate of *p*-process molybdenum isotopes, ^{92}Mo and ^{94}Mo, Lugmair, Scheinin, and Marti (1975) inferred $P_{146/144} = 0.57$. Audouze and Truran (1975), using solar seed abundances, give the first calculation of the production rate $P_{146/144}$ from an explosive nucleosynthesis for temperature-density conditions of $T \sim 2 \times 10^9$ K and $\rho \sim 10^4$ g cm^{-3}. From a *p*-process, "sensu stricto," they estimate $P_{146/144} \sim 1$. Woosley and Howard (1978) presented another calculation of nucleosynthesis of the proton-rich isotopes, called the γ-process by these authors to account for production by photodisintegration. The *p*-process, as defined by B^{2}FH and Audouze and Truran (1975) dominantly due to (p, γ) reactions, was considered by Woosley and Howard (1978) to be negligible because of the possibly astrophysically implausible conditions of temperature and density used by the earlier workers. Woosley and Howard (1978) estimated $P_{146/144} = 0.024$, which differs drastically from the previous *p*-process estimate and results in a very small amount of ^{146}Sm available at the beginning of the solar system, for the case of a continuous nucleosynthesis, far below the abundance of ^{146}Sm determined by meteorite measurements. The calculations of Woosley and Howard (1978) were done at a time when there was no evidence for ^{146}Sm in the solar system (Lugmair, Scheinin, and Marti 1975), and, therefore, gave consistent results. The $P_{146/144}$ values of Woosley and Howard (1978) are compatible with the present measurements only in the case of a predominant late spike (producing at least half of the ^{146}Sm and ^{144}Sm). This would imply that this γ-process occurs in completely different conditions than the other nucleosynthetic processes, since such large contributions of a terminal spike would induce other large isotope anomalies in other elements, which are not seen. Alternatively, the ^{146}Sm may be the product of an intense late-stage proton bombardment of dust by the early Sun as considered, for example, by Wasserburg and Arnould (1987) for the possible production of ^{26}Al and ^{53}Mn. In the case of a continous nucleosynthesis model, it is impossible to match our results with the production ratio of Woosley and Howard (1978). Our data are compatible with $P_{146/144}$ calculated by Audouze and Truran (1975). In this latter case, the observed amount of $^{146}\mathrm{Sm}/^{144}\mathrm{Sm}$ can be considered consistent with a continuous and uniform nucleosynthesis model without the need for a late spike. The divergent predictions of the *p*- and γ-processes for critical nuclides have also been considered by Clayton (1978) and Käppeler *et al.* (1982) who have favored the γ-process. A reexamination of the physical conditions, realistic for the *p*-process and γ-process nuclide syntheses, appears desirable, as well as the identification of a suitable astrophysical site.

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